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► **To cite this version:**

Lucas Massaro Sousa, Allan Pretti Ogura, Chayene Gonçalves Anchieta, Mathieu Morin, Nicholas Islongo Canabarro. Biomass Torrefaction for Renewable Energy : From Physicochemical, Bulk Properties, and Flowability to Future Perspectives and Applications. *Energy & Fuels*, 2024, 38 (19), pp.18367-18385. 10.1021/acs.energyfuels.4c03407 . hal-04771906

**HAL Id: hal-04771906**

**<https://ifp.hal.science/hal-04771906v1>**

Submitted on 7 Nov 2024

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# **Biomass torrefaction for renewable energy: from physicochemical, bulk properties and flowability to future perspectives and applications**

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**ABSTRACT**

18 Biomass and biomass wastes can be a source of renewable energy and fuels through valorization  
19 in thermochemical processes. Torrefaction is a thermal pre-treatment often employed for  
20 upgrading raw biomass. In addition to providing the status of current techniques used to  
21 characterize raw and torrefied biomasses (in terms of their flowability and physicochemical,  
22 thermal, and bulk properties), we comment on current applications with these bulk solids. The  
23 limitations of current characterization methods are also discussed with a view to future scopes  
24 with advanced techniques, particularly related to physicochemical properties. This review  
25 underscores a lack of systematic studies focused on the importance of comprehensive  
26 knowledge of raw and torrefied biomass properties to achieve better flowability, thereby  
27 contributing to more efficient and cost-effective industrial processes. Indeed, among the 1320  
28 literature papers evaluated in this review, 647 characterized the chemical/thermal properties of  
29 raw and torrefied biomasses, while 254 considered physical/bulk properties, and only 11  
30 assessed bulk solids' flowability. The scarcity of studies on flowability suggests that this  
31 parameter has not been considered important by most researchers for the demonstration of  
32 process feasibility. However, characterizing bulk solids flow behavior is critical for the proper  
33 design of handling equipment and ensuring smooth plant operation, thus minimizing risks  
34 associated with unforeseen expenses and prolonged time for process troubleshooting and  
35 equipment retrofit. Moreover, even when the flowability was measured in the 11 papers, it was  
36 difficult to compare results between studies as measurement techniques were not the same, nor  
37 were the biomass type or torrefaction conditions. This highlights the need for future research  
38 on the flowability of raw and torrefied biomass, intending to obtain more sound and broad  
39 conclusions for the flow behavior of such heterogeneous materials, culminating in the  
40 development of standardized protocols to improve biomass handling and processing at an  
41 industrial scale. Besides, most of the studies available in the literature were based on small-  
42 bench torrefaction units, producing only a few grams of torrefied biomass. This is generally a  
43 limiting amount of material for complete assessment of flowability at different bulk conditions,  
44 as well as to evaluate other important handling aspects at industrial scale, such as bulk solid  
45 segregation, quality of fluidization, and so on. In future works, we also suggest evaluating other  
46 heterogeneous feedstocks, such as municipal wastes or refuse-derived fuels, and performing a  
47 complete characterization for the bulk solids to facilitate technical decision-making in  
48 bioenergy and biofuels processes.

49

50 **Keywords:** Biochar, pyrolysis, powder handling, Hausner ratio, angle of repose, shear cell,  
51 fluidization.

## 52 **1 Introduction**

53 The expansion of agricultural production and industrial processing can lead to an increase  
54 in organic waste generation. In such a scenario, the valorization of biomass resources can  
55 contribute to a circular economy by considering biomass's potential reuse or processing from  
56 several feedstocks, including wood and forestry wastes, organic wastes, and agricultural  
57 residues. The demand for sustainable energy solutions has directed significant research  
58 attention towards biochar or torrefied biomass, a carbon-rich material produced from biomass  
59 through processes like torrefaction.<sup>1-4</sup>

60 Biomass residues are sustainable resources with large availability and easy access.  
61 However, its use in raw form for thermochemical conversion processes is generally  
62 compromised due to issues related to bulk solids handling and reduced energetic efficiency.<sup>5</sup>  
63 This is a consequence of raw biomass properties, consisting of low bulk density, high moisture  
64 content, reduced flowability, low calorific value, low energy yield, and high power  
65 consumption for grinding. To make biomass profitable for large-scale operations, a  
66 pretreatment step such as torrefaction is often necessary.

67 Figure 1 presents a simplified diagram describing dry biomass torrefaction. This partial  
68 thermal conversion occurs for mild temperatures (generally within 200°C and 300°C), in the  
69 absence of any oxidizing agents, and leads to the formation of three main products: volatile  
70 matters either (i) condensable (steam and primary tars) or (ii) non-condensable (mainly CO and  
71 CO<sub>2</sub>) and (iii) a solid brown to dark residue called torrefied biomass. Note that other treatments  
72 employing either “wet” or “steam” atmospheres can be also classified as torrefaction to obtain  
73 an upgraded solid fuel, as described elsewhere<sup>6,7</sup>.

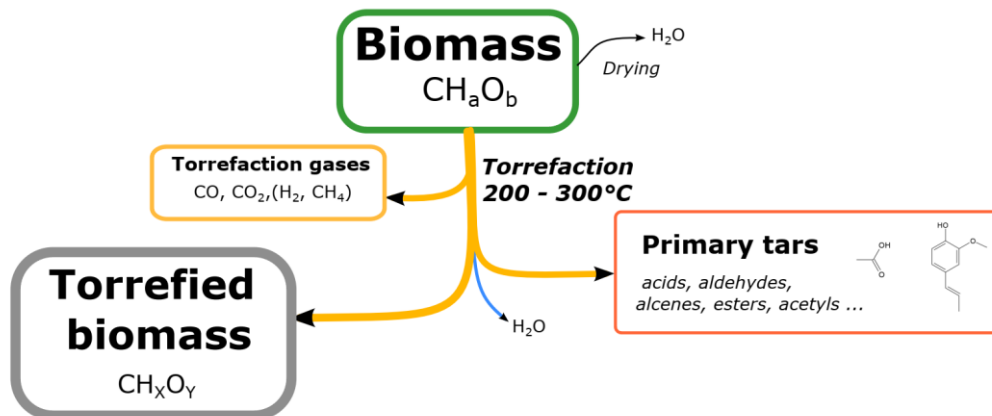


Figure 1. Diagram of dry biomass torrefaction.

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77 Industrially, two types of technology exist for dry biomass torrefaction according to the  
78 heating method<sup>8</sup>. In the first type, the heat can be supplied to the reactor by direct contact  
79 between hot inert gas and solids. This process mainly includes rotary drums, fluidized bed  
80 reactors, and multiple hearth furnaces. In the second type, the heat can be applied indirectly to  
81 the oven, by circulating, for instance, through a jacketed reactor. This process mainly includes  
82 screw reactors and rotary drums.

83 The torrefaction severity (or degree of torrefaction) is often characterized by the solid  
84 mass yield on a dry basis also called Anhydrous Weight Loss (AWL). For a given torrefaction  
85 technology and particle size, it mainly depends on two operating conditions: the temperature  
86 and the solid residence time. Indeed, a rise in these two parameters increases the degree of  
87 torrefaction which leads to the formation of a browner and darker torrefied solid. For instance,  
88 a typical mass yield for biomass torrefaction is 70% corresponding to an AWL of 30%.<sup>9,10</sup>  
89 According to the AWL value, it is then possible to i) significantly decrease the required energy  
90 for grinding since the higher the AWL, the lower the energy required for grinding<sup>11,12</sup>; ii)  
91 enhance fluidization and flowability characteristics due to the improvement of particle size and  
92 morphology, leading to more efficient handling operations;<sup>10</sup> iii) produce a hydrophobic solid  
93 with a low moisture content leading to long storage capacity<sup>13</sup>; iv) increase the biomass  
94 durability, which decreases the biological conversion and leads to less decomposition risks;<sup>8</sup> v)

95 increase the energy density with lower H/C and O/C ratios as the heating value is also increased  
96 to approach that of coal<sup>2,14</sup>.

97 Physical and chemical properties of torrefied biomass play a crucial role in determining  
98 its suitability for many applications. For instance, the versatility of torrefied biomass extends  
99 to energy applications, where it serves as a bridge between biomass and high-value products.  
100 In energy conversion, the high carbon content and stability of torrefied biomass make it a  
101 valuable precursor for producing activated carbon and other carbon-based materials. Its  
102 potential in energy storage is highlighted by its application in supercapacitors and batteries,  
103 where its porous structure and high conductivity are advantageous. Furthermore, the torrefied  
104 biomass can be used as a fuel, either as a direct combustion fuel or as a feedstock for producing  
105 syngas, biofuels, and bio-oil through gasification and pyrolysis.

106 Bulk and flowability characteristics of torrefied biomass are key for continuous operation  
107 and process yield. According to Dai *et al.*,<sup>15</sup> feeding waste biomass powders into  
108 thermochemical reactors is commonly the most problematic operation of the entire reactor  
109 system, because of blockages of pipes, feeding devices, and silo outlets. Issues arising from  
110 poor powder flowability can occur under a range of flow and stress conditions. For example,  
111 powder storage and discharge from silos often involves a quasi-static consolidated  
112 configuration for the powders, and common flow issues consist of irregularity and interruption  
113 of the flow of powders, rathole formation in funnel flow that can induce cross-contamination  
114 and rotting of material accumulated in the silo, slip-stick flow that can endanger the silo, etc.  
115 In contrast, processes such as fluidization and pneumatic transport involve a more dynamic and  
116 loose packing condition<sup>16</sup>, for which typical flow issues consist of blockage of pipelines,  
117 segregation and elutriation of finer particles, cross-contamination and rotting of accumulated  
118 material, etc. Therefore, different flowability methods and indices have been developed over  
119 the years to assess the flow behavior of a powder under different conditions of particle's

120 concentration, particle's relative movement, and their interaction with the system's boundaries  
121 (*i.e.*, if free-surface or confined systems). A complete discussion in this sense is provided in  
122 Section 3.3.1. Nevertheless, flowability data is key for the proper design of plant equipment  
123 (*e.g.*, silos, solid feeders, mechanical/pneumatic transfer lines, etc.) and can be useful to ensure  
124 smooth plant operation (*i.e.*, by monitoring flowability during operation as a quality parameter  
125 to avoid flow instabilities and equipment blockage if bulk solid properties change), avoiding  
126 unforeseen expenses and prolonged time for process troubleshooting and equipment retrofit.  
127 However, these flowability properties are often mistakenly overlooked in research publications,  
128 as if they were not necessary for demonstrating process feasibility.

129 Thus, this review paper aims to systematically evaluate the current state-of-the-art of  
130 biomass torrefaction, with a focus on the methods used for the characterization of raw and  
131 torrefied particulates, while proposing new fields of work based on our findings for advancing  
132 sustainable energy solutions. Also to provide the significance of understanding the complete  
133 properties of bulk materials, including their physical, chemical, thermal, bulk, flowability, and  
134 fluidization characteristics. There are currently some reviews on the physical and chemical  
135 characterizations of raw and torrefied biomass<sup>9,13,17,18</sup>; however, none focused on the  
136 characterization of flowability which is investigated in depth in our paper. The absence of  
137 comprehensive reviews on flowability is a critical knowledge gap that must be addressed,  
138 intending to guide future work that could lead to more efficient handling of biomass/biomass  
139 waste, reduce waste, and improve industrial processing methods. In this paper, the discussion  
140 is structured to cover several aspects of biomass torrefaction, as follows:

- 141 - The methodology used in this systematic review is presented in Section 2 and used in  
142 Section 3.1 to obtain quantitative information on the number of papers published,  
143 types of biomasses considered, and number of papers that investigated raw/torrefied

144 biomass characterization based on i) chemical & thermal properties, ii) physical &  
145 bulk properties, and iii) flowability & fluidization properties.

146 - In Section 3.2, we describe briefly the current methods used for the physicochemical  
147 characterization of raw and torrefied biomasses, while proposing other advanced  
148 techniques for future work to fill some knowledge gaps.

149 - In Section 3.3, we present an in-depth analysis of the flowability and fluidization of  
150 raw and torrefied biomasses, which is the core focus of our work. Flowability methods  
151 are critically reviewed and key information about torrefaction conditions and  
152 flowability data are extracted from the literature papers. The data is further grouped  
153 for visualization and critically analyzed on which methods and biomass types are most  
154 used, most productive countries, etc. Finally, we suggest future work to fill knowledge  
155 gaps on flowability and fluidization linked to biomass torrefaction.

156 - In Section 3.4, we describe the main applications with torrefied biomass extracted  
157 from the literature along with potential future trends.

158 - In Section 4, we propose final considerations on current and future literature, based  
159 on the critical analysis provided in this article and our findings.

160

## 161 **2 Systematic review methodology**

162 The systematic review consisted of three stages and followed the PRISMA protocol  
163 (Preferred Reporting Items for Systematic Reviews and Meta-Analyses),<sup>19</sup> considering the  
164 Scopus and Web of Science databases. As authors in the literature adopt different  
165 nomenclatures for the biomass after the torrefaction process, the chosen search string was  
166 “(‘biochar’ OR ‘biocoal’ OR ‘torrefied biomass’) AND ‘torrefaction’.” The research included  
167 documents in English, and it was current as of March 20<sup>th</sup>, 2024. In the first stage, documents  
168 from both databases were retrieved, and duplicates were removed. The gathered information



169 included title, publication year, authors, affiliation, and keywords. For each database, co-  
170 occurrence clusters (network visualization) were prepared in VOSviewer® software (version  
171 1.6.19) with the keywords of the selected articles.

172 In the second stage, there was a selection of papers within the scope of our research that  
173 focused on biomass torrefaction, including some wastes that contain biomass, such as municipal  
174 solid wastes, refuse-derived fuels, or solid recovered fuel. It was considered as out of scope  
175 non-experimental research such as reviews and simulations as they do not provide new  
176 information about bulk solid properties or characterization methods. For the selected papers,  
177 we retrieved information regarding the type of biomass used for torrefaction to provide an  
178 overview of the most studied feedstocks and their relevance in the context of a circular  
179 economy. The remaining papers underwent individual qualitative analysis, categorizing them  
180 as “yes”, “limited”, or “no” based on the availability of information about the physical,  
181 chemical, and flowability characteristics of raw and torrefied biomass. Basic chemical  
182 characterization should include proximate analysis, such as moisture, volatile matter, fixed  
183 carbon, and ash contents, and ultimate analysis, involving the contents of carbon, hydrogen,  
184 oxygen, nitrogen, sulfur, and other elemental compositions. In addition, the provision of  
185 thermal characteristics should also be considered, such as mass and energy yields and heating  
186 values. Physical and bulk characteristics involve particle size distribution, true and bulk density,  
187 porosity, specific surface area, and morphological features, such as shape observed through  
188 techniques such as scanning electron microscopy (SEM). Flowability characteristics include  
189 static and dynamic angle of repose, Hausner ratio, Carr index, and flow function measured via  
190 rheometers. Additionally, information on fluidization curves and permeability was also  
191 considered.

192 In the third stage, we focused on studies that have conducted experimental approaches to  
193 determine flowability characteristics. Additionally, we included studies from other reviews or

194 those cited within the selected papers that were relevant to our research scope but were not  
195 retrieved in our systematic process. Flowability assessment of blends between torrefied biomass  
196 and pulverized coal for co-processing operations was considered out of the scope of this review  
197 paper, particularly if the flowability characterization was not performed for the raw biomass.  
198 Some interesting papers on this subject can be found elsewhere.<sup>20-22</sup> For the final selection of  
199 papers, we have provided information on the flowability or fluidization of torrefied biomass,  
200 discussed their main characteristics and critically reviewed these studies. Finally, we explored  
201 current applications of torrefied biomass and future perspectives, offering suggestions for future  
202 research.

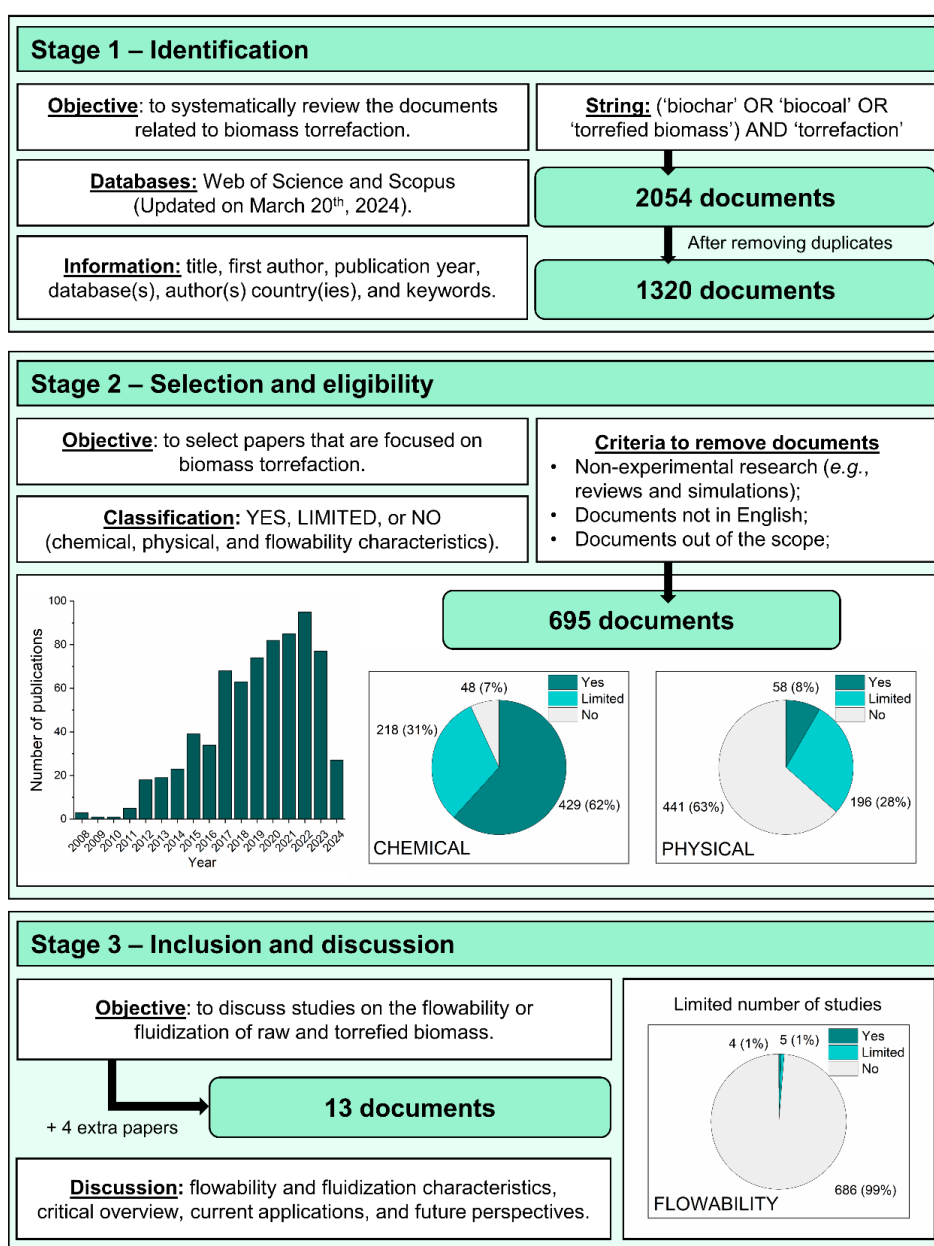
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### 204 **3 Results and Discussion**

#### 205 3.1 Results of systematic review

206 Figure 2 presents a summary of the systematic review process and the selection of papers  
207 for each stage. In the first stage, 2054 documents were retrieved, with 1143 from Web of  
208 Science and 911 from Scopus. Figures S1 and S2 (Supplementary Information) present the co-  
209 occurrence clusters with the keywords and affiliations of the selected articles from each  
210 database. After removing duplicates, 1320 documents remained. In the second stage, 695  
211 documents were selected based on our eligibility criteria. Considering the number of  
212 publications per year, we observed an increasing trend, which aligns with the emerging interest  
213 in this field of study. For instance, most papers (58%) were published in the last five years (74  
214 in 2019, 82 in 2020, 85 in 2021, 95 in 2022, and 77 in 2023). Most papers presented complete  
215 chemical characteristics of the torrefied biomass (62%) compared to those that presented  
216 physical characterization (8%). More than half of the papers (63%) did not provide sufficient  
217 physical and bulk characteristics of the torrefied biomass, which is a limitation for the

218 predictability and optimization of industrial operations with these materials. Additionally, the  
 219 different characterizations provided in each research challenge the comparison across studies.  
 220 A brief overview of the chemical and physical characterization is presented in Section 3.2.  
 221 Finally, the systematic approach retrieved 9 papers that assessed the flowability or fluidization  
 222 of raw and torrefied biomass. Additionally, 4 documents were found either within the selected  
 223 paper's bibliography or in other reviews. Therefore, 13 studies were selected and critically  
 224 discussed in Section 3.3.



225  
 226 Figure 2. Systematic review summary: The search process and selection of the studies focused  
 227 on the flowability of fluidization of raw and torrefied biomass.

228 From all papers selected in the second stage (n = 695), wood-derived biomass was  
229 identified as the most common feedstock for torrefaction (n = 326). These papers reported the  
230 repurposing of wood chips, pellets, sawdust, shavings, and stems. Some studies compared the  
231 biomass torrefaction of softwood (*e.g.*, pinewood) and hardwood (*e.g.*, teakwood).<sup>23</sup>  
232 Additionally, other studies reused waste wood from furniture manufacturing as a recycling  
233 alternative.<sup>24</sup> Several factors explain the predominance of studies on wood, including its  
234 abundance and availability, high energy density, and homogeneous composition. Several  
235 varieties of grasses and herbaceous plants were notable among the studies (n = 67), including  
236 pruning and weeding residues, with *Miscanthus* spp. standing out (n = 11).

237 Agro-industrial residues constitute a significant group in the selection of feedstocks for  
238 biomass torrefaction, contributing to a circular economy through waste minimization and the  
239 repurposing of by-products from industrial processes.<sup>25</sup> Rice residues, including husks and  
240 straws, are extensively studied (n = 65), followed by corn stalks, stover, straw, and corncob (n  
241 = 55), and wheat straw (n = 30). These feedstocks are among the three largest cereal crops  
242 worldwide,<sup>26</sup> ensuring abundant residues available for biomass torrefaction. Sugarcane bagasse,  
243 leaves, and straw (n = 28) also play a crucial role in enhancing the energy balance of agro-  
244 industries and supporting ethanol production. Additionally, residues such as shells and husks  
245 from various nuts (*e.g.*, almonds, coconuts, walnuts, hazelnuts, and peanuts) are studied (n =  
246 27). Other agricultural biomass includes oil palm trunk, kernel shells, mesocarp, and fronds (n  
247 = 31); coffee residues such as spent grounds and husks (n = 23); cotton stalks (n = 14); olive  
248 stones, cake, kernel, pomace, pips, and oil residue (n = 14); sunflower seeds, husks, and stalks  
249 (n = 10). Barley (n = 6), camellia (n = 6), oats (n = 4), canola (n = 3), cassava (n = 3), pigeon  
250 pea (n = 3), and soybean (n = 3) residues have also been studied.

251 Specific organic wastes such as fruit peels, seeds, shells, and stones, along with empty  
252 fruit bunches have been studied for biomass torrefaction (n = 18). Municipal solid waste,

253 particularly its organic fraction, has been studied in 10 papers, and the torrefaction process can  
254 produce a high energy density solid fuel.<sup>27</sup> Sewage sludge (n = 18), microalgae (n = 20) such  
255 as *Chlorella vulgaris*, mushrooms (n = 7), refuse-derived fuels (n = 6), and textile waste (n =  
256 4) are unconventional but efficient feedstocks for torrefaction. Poultry and chicken manure (n  
257 = 15), as well as cattle manure (n = 7), are livestock residues that have been studied for  
258 torrefaction. Additionally, 54 papers have explored other types of biomasses, including  
259 polyamide biocomposites, distillation residues, tea residues, among others.

260

### 261 3.2 Chemical and physical characterization

262 During torrefaction, biomass undergoes several transformations, leading to some  
263 chemical and physical modifications. First, the moisture is removed by the initial drying step  
264 (up to 150°C), the second stage remains relatively stable within the temperature range of 137°C  
265 –181°C, to remove chemisorbed water, followed by chemical transformations of the  
266 hemicellulose, cellulose and lignin for temperatures range of 180°C up to 580°C, and releasing  
267 volatile compounds and solids. The produced torrefied biomass is an upgraded solid that is an  
268 alternative to fossil fuels<sup>28</sup> for energy storage and conversion systems.<sup>29,30</sup> The torrefaction  
269 process increases the calorific value of the biomass, resulting in a more efficient fuel. As the  
270 torrefied biomass is easier to be ground into a fine powder, it could also be used as a pulverized  
271 fuel. Torrefaction significantly enhances the physical and chemical properties of biomass,  
272 transforming it into a more efficient and versatile fuel source.

273 Through detailed physical and chemical characterization, the quality and suitability of  
274 biomass torrefaction for various applications can be assessed and optimized, promoting its  
275 adoption in sustainable energy and industrial systems. While there is substantial literature on  
276 biomass characterization, this section focuses on presenting the techniques used for such  
277 characterization and provides a critical perspective on the existing gaps in information.

278           Although the chemical and thermal characterization are available in 647 papers  
279 screened in this study, there are limited papers which present both chemical/thermal and  
280 physical/bulk characterization. For this purpose, the systematic review procedure selected 58  
281 papers that have analyzed the physical and bulk properties properly (Table S1), in which 36  
282 papers (Table S2) include chemical and thermal characterization (see these tables in the  
283 Supplementary Information). The objective of the next subsections is to describe the trends of  
284 characterization methods used for torrefied biomass, in terms of chemical and thermal  
285 properties (Section 3.2.1) as well as physical and bulk properties (Section 3.2.2).

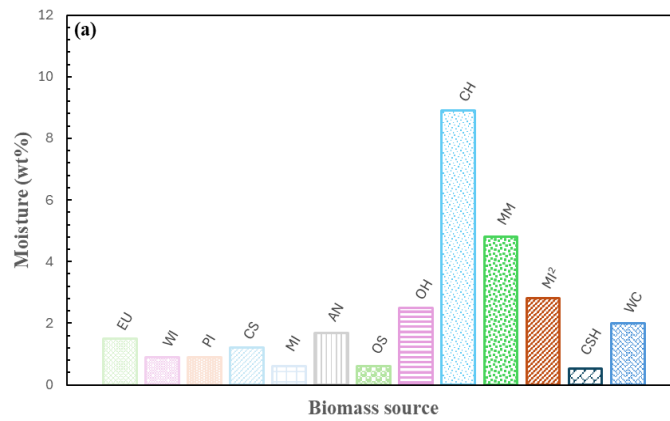
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### 287 3.2.1 Chemical and thermal characterization of biomass and torrefied biomass

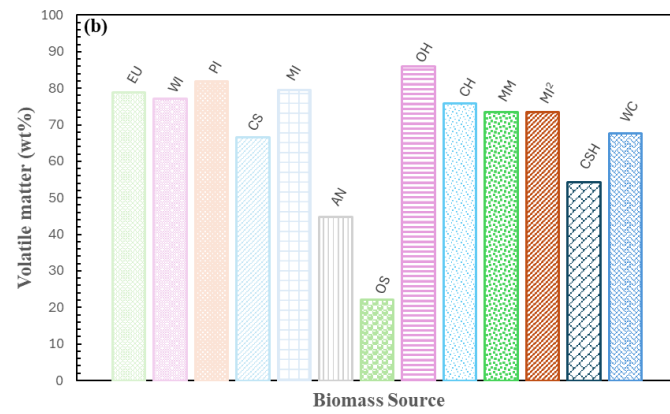
288           Chemical changes could be addressed by focusing on the changes in chemical  
289 composition or/and structure. The proximate analysis involves determining the moisture,  
290 volatile matter, fixed carbon, and ash content. The torrefied biomass may present a significant  
291 discrepancy in the proximate analysis results, considering the different methods, such as those  
292 from the American Society for Testing and Materials (ASTM) and thermogravimetric analysis  
293 (TGA), and the process parameters (*e.g.*, heating rate, gas flow, temperature) used to obtain it.  
294 The elemental composition related to carbon, hydrogen, nitrogen, sulfur, and oxygen  
295 (CHNS/O) supports searching for elemental changes along the torrefaction process, such as the  
296 amount of carbon which an increase is expected, enhancing the material's energy density<sup>6,7</sup>.  
297 The amount of CHNS/O may vary according to the source of biomass, and the ultimate analysis  
298 is important for understanding biomass thermochemical conversion. Although the chemical  
299 characterization plays a role, the intrinsic nature and recalcitrance<sup>31</sup> of the biomass leads to a  
300 considerable variation for ultimate and proximate analyses (see Table S2). The values vary from  
301 biomass to biomass and even within the same type of biomass. Figure 3 addresses some

302 important information to illustrate the torrefied biomass composition and to highlight this  
 303 statement.

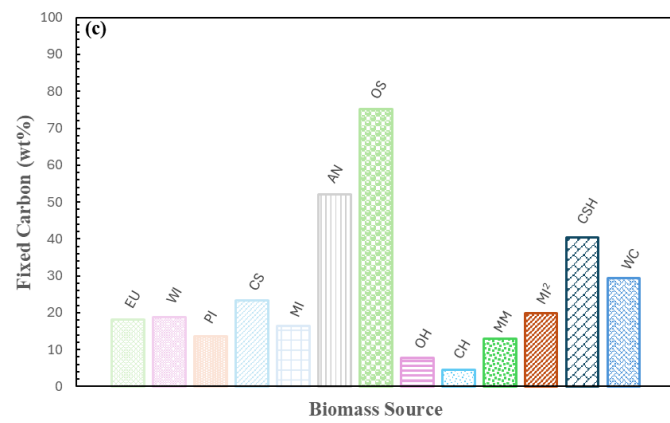
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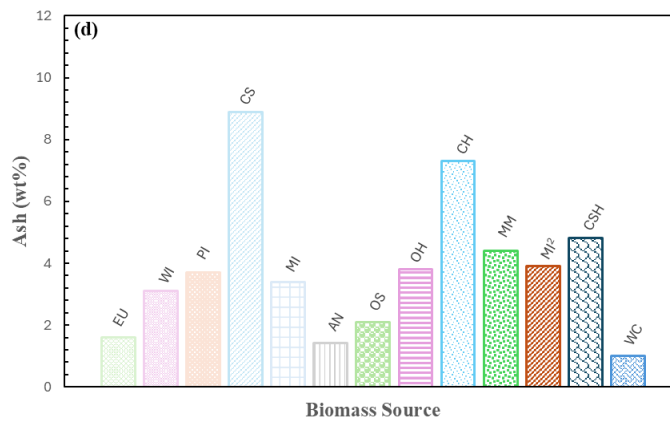
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307



308 Figure 3. Proximate analysis of different biomass sources. (EU: Eucalyptus, WI: Willow, PI:  
309 Pine, CS: Corn Straw, MI: Miscanthus, AN: Acacia nilotica, OS: Olive stone, OH: Oat hull,  
310 CH: Canola hull, MM: Mustard meal, MI2: Miscanthus, CSH: Camellia shell, WC:  
311 Woodchips). (For reference see Table S2)<sup>32-37</sup>  
312

313 The heating value of torrefied biomass is higher due to an increase in the carbon content  
314 and due to the reduction of moisture. This could be evidenced by the TGA in which the stability  
315 and decomposition are presented and defined by mass losses. TGA characterization can provide  
316 insights into the combustion and pyrolysis kinetics, moisture, biomass degradation, endo and  
317 exothermic reactions, ash content, which gives an overview of biomass behavior. The thermal  
318 analysis will help to define future applications with torrefied biomass.

319 Another important characterization is Fourier Transform Infrared Spectroscopy analysis  
320 (FTIR), which is used to identify changes in functional groups and chemical bonds, revealing  
321 the extent of thermal degradation and the formation of new compounds. Some physicochemical  
322 characterization, such as X-ray-based (*e.g.*, synchrotron-based scattering (SAXS), tomography,  
323 Diffraction (XRD), Photoelectron Spectroscopy (XPS), fluorescence (XRF), Computed  
324 Tomography (CT), etc.), need to be better defined in the context of torrefied biomass.<sup>38-40</sup>  
325 Among the papers selected in this study (see Table S2), only five<sup>33,37,38,41,42</sup> explored this type  
326 of characterization to understand torrefied biomass. XRD provides a deep understanding of the  
327 crystalline structure and can indicate the changes in the biomass's structural properties post-  
328 torrefaction. XRD is important to characterize the type of carbon obtained during the process  
329 (*e.g.*, graphite carbon atoms are arranged in layers, while graphene is only a single layer of  
330 carbon atoms).<sup>43</sup> Combining the XPS technique with Raman Spectroscopy, it could be possible  
331 to provide information regarding the type of carbon chemical bonds (*e.g.*, C=O, C-C, C-O) and  
332 is possible to provide information about carbon defects, which could interfere with the energy  
333 technologies applications. A full understanding of carbon sites could facilitate its application in  
334 the context of energy technologies.<sup>44,45</sup>



335 Nuclear Magnetic Resonance (NMR) has been used to provide detailed information about  
336 the local chemical environment of carbon atoms, including the types of carbon-carbon bonds,  
337 the presence of functional groups, and molecular structure<sup>46</sup>. It can distinguish between  
338 different types of carbon atoms, such as  $sp^2$ ,  $sp^3$ , and hybridized carbons. However, this  
339 technique has some disadvantages as it requires relatively large sample amounts, and the  
340 analysis can be time-consuming. It is less effective for highly crystalline materials because it is  
341 more focused on the local environment rather than long-range order.

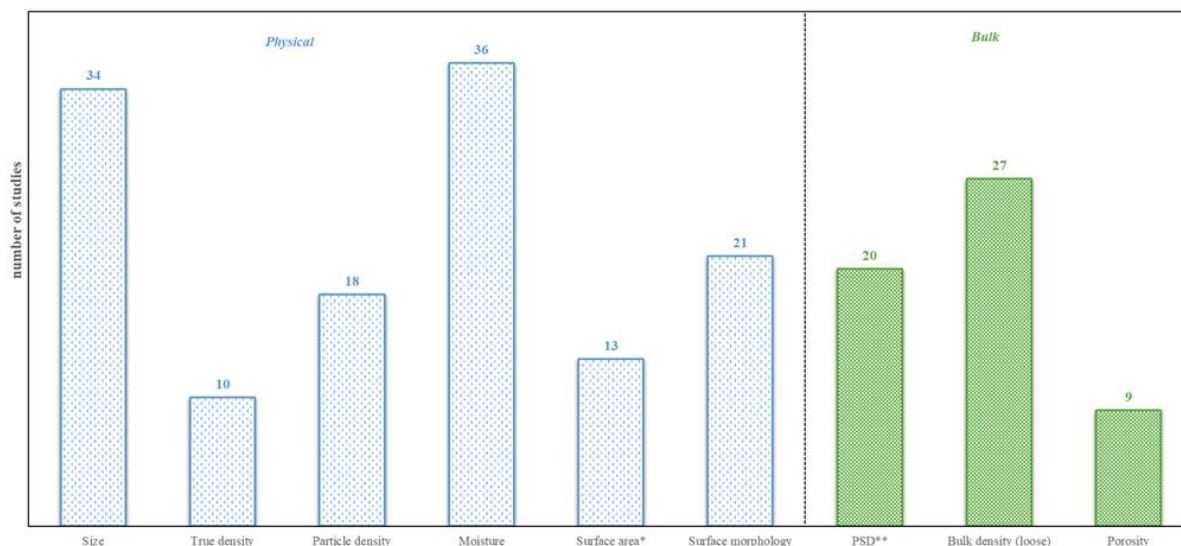
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### 343 3.2.2 Physical and bulk characterization of biomass and torrefied biomass

344 Knowledge about particle characteristics is important for the design of processes and  
345 helps further optimizations. In terms of properties, it is needed to investigate those related to a  
346 single particle, but also those that account for many particles and the voids within, known as  
347 bulk properties. Overall, raw and torrefied biomass and biomass wastes present considerable  
348 heterogeneity of particle and bulk properties, which is linked to different biomass types, nature  
349 and structure but also to bulk solids processing methods and process conditions.

350 From the systematic review, 58 papers were selected with an appropriate characterization  
351 of physical and bulk properties (Figure 4). In these papers, the most investigated biomass for  
352 torrefaction is woody-derived ( $n = 30$ ), followed by agro-industrial residues ( $n = 23$ ),  
353 wastewater ( $n = 2$ ) and algae ( $n = 1$ ), respectively. It is worth noting that some researchers  
354 evaluated the impact of torrefaction from distinct sources of biomass. For this reason, the total  
355 number of articles cited above exceeds the number of selected studies. Besides biomass  
356 heterogeneity, the methods for biomass characterization are also a relevant factor that may lead  
357 to some discrepancies in the measured properties values. For instance, 21 studies investigated  
358 the particle size distribution (PSD) of raw and torrefied biomass, respectively. Among those,  
359 18 studies assessed PSD by sieving out the particles and applying a gravimetric method while

360 fewer use laser diffraction ( $n = 2$ ) and image analysis ( $n = 1$ ). Different experimental procedures  
 361 may result in significant differences in particle size distribution, which makes comparison  
 362 between studies sometimes challenging.



363  
 364 **Figure 4. Physical and bulk characterization of studies englobing biomass torrefaction.**  
 365 (\*Surface area is related to BET measurements, and it also involves mean pore size and pore  
 366 volume estimative; \*\* Particle size distribution. (For reference source see Table S1).  
 367

368 Bulk density is also a critical property since it mainly impacts process storage and  
 369 handling. Within the selected papers in this study ( $n = 58$ ), 27 papers estimated values for bulk  
 370 density for raw and/or torrefied biomass. 22 papers estimated values for loose bulk density and  
 371 3 papers estimate values for tapped bulk density. Moreover, only 2 papers estimated values for  
 372 both loose and tapped bulk density. There is often a misunderstanding between the definition  
 373 and use of terms such as loose bulk density and tapped bulk density. The first one refers to the  
 374 not compressed state, while the second one is the density of the material after it has been  
 375 compacted.<sup>47,48</sup> Considering the heterogeneous particle size, the shape of the biomass and the  
 376 process conditions (*e.g.*, pressure), clarifying this distinction is essential for handling it over  
 377 upstream and downstream processes. Loose and tapped bulk densities are also important for  
 378 estimating flowability parameters such as Hausner index and Carr's compressibility factor,  
 379 which are presented in the section 3.3.1.

380 Aligned with the discussion above, it is clear that bulk and physical properties should be  
381 assessed carefully, and the methods adopted to estimate these properties must be in line with  
382 the process phenomena. Bulk and particle properties affect flowability, which is an important  
383 parameter for efficient handling and storage of bulk solids in different applications, particularly  
384 for bioenergy and biofuels.

385

### 386 3.3 Flowability and fluidization characterization of biomass and torrefied biomass

387 From the systematic review procedure, a total of 13 papers were obtained to be critically  
388 evaluated concerning the effect of torrefaction on biomass flowability characteristics ( $n = 11$ )  
389 and fluidization characteristics ( $n = 2$ ). The most used feedstock consisted of wood-derived  
390 biomass ( $n = 8$ ) followed by agro-industrial residues ( $n = 4$ ), and one single occurrence for  
391 waste textile fiber. The fluidization papers were published from 2012 to 2013, whereas those  
392 addressing the flowability characteristics were published more recently, from 2017 to 2023.  
393 With respect to flowability, the most productive countries were India and France with 4  
394 occurrences each, followed by single occurrence papers from China, Mauritius, and Philippines.  
395 Interestingly, among these papers the affiliation of the authors sometimes varied but it was  
396 always within the same country. Scientific collaborations with experts from different  
397 backgrounds in different countries should be promoted in the future. Finally, all the occurrences  
398 for the fluidization papers were from Brazil ( $n = 2$ ) with one co-author from France in one  
399 paper.

400 In the following subsections, the flowability methods (Section 3.3.1) and selected papers  
401 (Section 3.3.2) are critically reviewed, this latter in terms of main findings, torrefaction  
402 conditions, type of bulk solid and flowability results interpretation. The flowability data from  
403 all the papers are further grouped in Section 3.3.3 to discuss their trends and limitations. Finally,  
404 a similar assessment is done for the selected fluidization papers in Section 3.3.4.

405 3.3.1 *Definition of flowability and critical overview of current methods*

406 By definition, a bulk solid is composed of solid particles and voids that are surrounded  
407 by the environment or system boundary. The particles can have different physicochemical  
408 characteristics (*e.g.*, various shapes, densities, sizes, chemical compositions, surface textures,  
409 etc.), whereas the interstitial voids can be filled with different fluids of different densities and  
410 viscosities. The environment or system boundary can have different arrangements, such as rigid  
411 or moving walls of different textures, freeboards in which the bulk solid can be in contact with  
412 fluids, or even regions of different pressures and temperatures. From this rationale, it is clear  
413 that the environment impacts the bulk solids arrangement, and that its interaction combined  
414 with the particularities of particles and voids characteristics, creates a complex system, which  
415 is one of the reasons that developing a universal index to describe “flowability” is a challenging  
416 task.

417 Bulk solid flowability can be defined as the likelihood of a particulate material to flow,  
418 or in other words, as the potential for the particles to overcome the bulk inertia and have a  
419 relative movement over other particles and its surroundings. There are different forces that  
420 could prevent particles from moving, which can be of electrostatic origin, cohesive origin, or  
421 even mechanical/interlocking<sup>49</sup>. These have been well documented in the literature, although  
422 direct measurement of these forces is still a challenge.<sup>50,51</sup> Over the years, several indices were  
423 proposed to assess, quantify, and discriminate the flowability of bulk solids, which have  
424 different complexities but are fundamentally based on using a limited amount of material to  
425 infer about the flow behavior of the bulk for a given process, generally at a much higher scale.  
426 A critical review of current methods for flowability assessment is presented below.

427 Some classical methods involved the evaluation of flowability through simple powder  
428 compaction tests, in which the degree of variation of the bulk density, between loose and  
429 tapped/consolidated conditions, was used to indicate a better or worse flow behavior. In short,

430 powders that show a higher difference between these two conditions are classified with a worse  
431 flowability, which is attributed to a higher intensity of electrostatic, cohesive, and/or  
432 interlocking forces. The Hausner ratio (HR) is an index given by the ratio between  
433 tapped/consolidated and loose conditions, whereas Carr's compressibility index (CCI) is given  
434 by the standard difference between these variables.<sup>52,53</sup> Powder compaction can be done by  
435 vibration using automated or manual devices, for which the bed weight is the driven force for  
436 bulk compaction, or even by using devices that impose consolidating pressures, in which there  
437 is generally higher compaction of the powder bed. One important remark from these indices is  
438 that they are measured under conditions of considerable high particle fraction, approaching  
439 powders' packing limit; hence, the qualitative interpretation of the flowability might be  
440 extrapolated, at best, to processes that undergo similar traits. Nonetheless, these indices are  
441 mostly known for their quick and easy measurement that can be useful for a qualitative  
442 differentiation between samples, and not necessarily for direct extrapolation to process  
443 conditions. Some interesting works on these indices are presented elsewhere, for example  
444 through modeling of the compaction procedure and via assessment of the impact of particle size  
445 distribution, moisture content, ambient humidity and temperature, and so on.<sup>47,54-60</sup>

446 Another classical index is the static angle of repose ( $\alpha$ ), which can be measured by the  
447 fixed height or fixed diameter methods. The bulk solid is discharged by gravity through a funnel  
448 to make a cone trunk on a flat or slightly concave surface. The literature rationale is that this  
449 slope angle of the powder bed can be associated with higher or lower intensity of cohesion  
450 forces. In short, a higher slope angle is an indication of worse flowability for the bulk solid.  
451 Some authors claim that this index can be correlated with particle properties or even  
452 dimensionless numbers, while others show that the angle of repose is only useful to plan storage  
453 in a pile for bulk solids or predict the inventory in silos.<sup>61,62</sup> It is worth noting, however, that  
454 this test is performed under free-discharge of powders and that the pile is formed under loose

455 bulk conditions without any applied consolidation; hence, any extrapolation of flowability  
456 information from this index should be done, at best, in such flowing conditions in the process.  
457 One of the main advantages of this index is that it can be measured quickly and easily and can  
458 give a first qualitative indication to discriminate the flowability of different samples,  
459 particularly those samples that have considerable differences from each other. Some interesting  
460 papers addressing the measurement of the angle of repose for bulk solids with different  
461 properties can be found elsewhere.<sup>48,62-65</sup>

462         The flow function coefficient (FFC) is another index that has been used for evaluating the  
463 flowability of bulk solids, which is defined as the ratio of the major consolidation stress to the  
464 unconfined yield strength. These parameters can be obtained in shear cells, which are devices  
465 where the bulk solid is subsequently consolidated and sheared to derive the flow function at  
466 each consolidation. As a consequence, the flowability indication can be evaluated at different  
467 bulk solid conditions, corresponding to processes for which the bulk solids flow at  
468 concentrations close to the packing limit and at different consolidation pressures. Note that this  
469 is a different feature than the previous static angle of repose and compressibility indices, which  
470 are measured for one condition generally at small bed weight consolidation. There are many  
471 different commercial implementations of shear cells, from the precursor Jenike shear cell to the  
472 Schulze ring shear tester, Brookfield powder flow tester, Freeman FT4 rheometer, and so on.  
473 These implementations have different pros and cons concerning the consolidation range,  
474 measurement speed, and degree of automatization. In general, the standard procedure of these  
475 pieces of equipment leads to results with low variability under well-defined conditions (*e.g.*,  
476 ASTM D6773-16).<sup>66</sup> Furthermore, the results of the shear cells allow not only for a qualitative  
477 differentiation between the flow behavior of particulates but also provide valuable information  
478 for the design of hoppers and silos since the cohesion, particle-particle, and particle-wall  
479 friction parameters can be obtained from the tests. The effect of environment conditions, and

480 particle properties on the flow function has been extensively investigated in the literature,  
481 including the comparison between the flowability indications given among the different  
482 commercial shear cells.<sup>67-73</sup> Some authors also commented on the limitations of shear cells to  
483 measure flow properties of elongated or fibers type of biomass materials that are highly  
484 compressible and severely entangled, which hinders the formation of a well-defined shear zone  
485 and the attainment of steady-state flow condition.<sup>74,75</sup>

486 More recently, rotating drums coupled with image analysis have been used to evaluate  
487 bulk solids flowability at dynamic conditions. According to some authors, this device measures  
488 dynamic flowability and can be understood as a shear cell at low consolidating stresses.<sup>76</sup> The  
489 particulate is introduced in a drum that is continuously rotated while different characteristics of  
490 the flow can be evaluated, mostly related to powder bed angle and the flow irregularity. It is  
491 worth noting that depending on the drum rotating speed, different flow regimes can develop,  
492 from unstable and intermittent to stable, cascading, cataracting, and avalanching modes, posing  
493 more challenges for determination compared to the quasi-static indices (*i.e.*,  $\alpha$ , HR, and FFC).  
494 Furthermore, there is a greater interaction between fluid and bulk solids during the measurement  
495 of this index, which can be challenging as the properties of solid and void fluid vary. For non-  
496 cohesive materials, the angle that the particulate material makes with the horizontal under stable  
497 flow is commonly named the dynamic angle of repose ( $\beta$ ). However, for cohesive materials like  
498 biomass, different parameters can be tracked between powder avalanches, such as the upper  
499 and lower angle of stability, the frequency of avalanches, and so on.<sup>16</sup> Generally, the higher the  
500 dynamic angle of repose or angle of upper stability, the higher the interparticle frictional forces  
501 in the powder, which is potentially an indication of worse flowability. The effect of bulk and  
502 particle properties on the dynamic angle of repose has been evaluated in some works, however,  
503 with considerably fewer data than shear cells.<sup>77-79</sup> An indicative classification between powder

504 flowability as a function of dynamic angle or other dynamic quantities is still under  
505 investigation.<sup>80,81</sup>

506 From the rationale described in the previous paragraphs, the technique for flowability  
507 assessment should be evaluated with caution and, more importantly, the test should be in line  
508 with the actual process phenomenon under investigation, in which the bulk condition from the  
509 process should be well reproduced, using a limited amount of material, during flowability tests  
510 to allow for results interpretation and extrapolation at different scales.

511

### 512 *3.3.2 Description of selected papers for flowability*

513 In this section, the focus is to describe the main findings of the 11 papers that investigated  
514 raw/torrefied biomass flowability and extract key information about torrefaction conditions,  
515 type of bulk solid, flowability methods employed, and results interpretation by authors. A  
516 chronological order is followed, when possible. The flowability data is summarized in Table 1,  
517 along with the biomass type, torrefaction temperature and residence time, type of torrefaction  
518 technology and unit scale, as well as other characterization methods used in each paper for  
519 obtaining chemical, physical, bulk, and thermal properties.



520 **Table 1.** Flowability indices for the selected papers, consisting of Hausner Ratio (HR), Carr's Compressibility Index (CCI), static repose angle  
 521 ( $\alpha$ ), dynamic repose angle ( $\beta$ ), and flow function (FFC) at different pre-consolidation loads.

Authors	Biomass	T (°C)	$\Delta t$ (h)	HR (-)	CCI (%)	$\alpha$ (°)	$\beta$ (°)	FFC <sup>a</sup> (-)	FFC <sup>b</sup> (-)	FFC <sup>c</sup> (-)	FFC <sup>d</sup> (-)	Other information
Conag et al. (2017) <sup>82</sup>	sugarcane bagasse (SB)	0	0	1.49	33.00							- Fixed bed torrefaction at bench scale (9g of material).  - Other tests performed: proximate analysis, calorific value, particle size, bulk density, combustion indices, and equilibrium moisture.
		250	0	1.44	30.33							
		250	0.25	1.42	29.33							
		250	0.5	1.32	24.00							
		250	0.75	1.32	24.00							
		250	1	1.25	19.67							
		250	1.25	1.33	24.67							
		300	0	1.44	30.33							
		300	0.25	1.32	24.00							
		300	0.5	1.31	23.67							
		300	0.75	1.37	27.00							
		300	1	1.39	28.00							
		300	1.25	1.35	25.67							
		350	0	1.35	25.67							
		350	0.25	1.27	21.33							
350	0.5	1.30	23.00									

		350	0.75	1.35	26.00	
		350	1	1.41	29.00	
		350	1.25	1.45	31.00	
Rago et al. (2018) <sup>83</sup>	waste textile fiber (WTF)	0	0	1.34	25.22	- Fixed bed torrefaction at bench-scale (65g).
		225	1	1.22	17.82	- Other tests performed: proximate and ultimate analyses, calorific value, FTIR, particle size, and bulk density.
		250	1	1.20	16.92	
		275	1	1.10	8.85	
		300	1	1.08	7.77	
		225	3	1.26	20.59	
		250	3	1.25	19.81	
		275	3	1.11	10.06	
		300	3	1.04	3.94	
Xu et al. (2019) <sup>84</sup>	fine soybean straw (SB)	0	0		60.9	- Fixed bed torrefaction at bench-scale.
		300	0.75		55.3	- Other tests performed: proximate analysis, SEM, particle size, PSD, bulk density, and shape factor.
	coarse soybean straw (SB)	0	0		61.2	
		300	0.75		55.3	
	fine corn straw (CS)	0	0		61.4	
		300	0.75		59.3	
	coarse corn straw (CS)	0	0		54.8	
		300	0.75		54.2	

	fine rice straw (RS)	0	0		64.9	
		300	0.75		56.8	
	coarse rice straw (RS)	0	0		70.0	
		300	0.75		68.2	
	fine rice husk (RH)	0	0		44.3	
		300	0.75		43.6	
	coarse rice husk (RH)	0	0		44.7	
		300	0.75		43.9	
Singh et al. (2019) <sup>85</sup>	pigeon pea stalk (PPS)	0	0	1.48	32.21	- Fixed bed torrefaction at bench-scale (6g).
		225	0.25	1.49	32.97	- Other tests performed: proximate and ultimate analyses, calorific value, FTIR, SEM, particle size, bulk density, combustion indices, and equilibrium moisture.
		225	0.5	1.46	31.55	
		225	0.75	1.44	30.65	
		250	0.25	1.39	27.93	
		250	0.5	1.38	27.77	
		250	0.75	1.38	27.66	
		275	0.25	1.38	27.75	
		275	0.5	1.37	27.31	
		275	0.75	1.36	26.51	
Singh et al. (2019) <sup>33</sup>	gum arabic wood (GAW)	0	0	1.29	22.48	- Fixed bed torrefaction at bench-scale (8g).
		252	1	1.21	17.35	- Other tests performed like previous work <sup>85</sup>

Singh et al. (2020) <sup>86</sup>	eucalyptus wood (EW)	0	0	1.44	30.43				- Fixed bed torrefaction at bench-scale (10g).	
		220	0.33	1.35	26.20				- Other tests performed: proximate and elemental analysis, calorific value, TGA, FTIR, XRD, BET, SEM, particle size, bulk density, particle density, combustion indices, and equilibrium moisture.	
		220	0.67	1.30	23.26					
		220	1	1.26	20.93					
		250	0.33	1.23	18.43					
		250	0.67	1.20	16.56					
		250	1	1.23	18.54					
		280	0.33	1.21	17.36					
		280	0.67	1.24	19.24					
		280	1	1.20	16.95					
Singh et al. (2020) <sup>87</sup>	gum arabic wood (GAW)	0	0	1.29	22.85	39.36			- Fixed bed torrefaction at bench-scale.	
		220	0.67	1.27	21.44	38.79			- Other tests performed like previous work <sup>86</sup> except for XRD, BET, but including contact angle, and ICP-MS.	
		250	0.67	1.21	17.35	36.15				
		280	0.67	1.18	15.86	32.51				
Pachon-Morales et al. (2019) <sup>88</sup>	poplar wood (PW)	0	0				5.2	4.6	3.9	- Fixed bed torrefaction at bench-scale (15g).
		220	1				5	4.4	4.2	- Other tests performed: PSD, shape factor, and bulk density.
		220	5				4.8	4.9	4.1	
		250	1				5.3	5	5.1	
		250	5				6.9	6	6.3	
		280	1				6.6	6.3	5.7	

		280	5		9.2	9.4	8.5		
	spruce wood (SW)	0	0		3.5	3.9	3		
		220	1		3.7	3.8	3.4		
		220	5		4.9	4.5	4.1		
		250	1		4.7	4.8	4.2		
		250	5		6.5	7.4	6.2		
		280	1		6	6.2	6.3		
		280	5		5.8	8.4	11		
Pachon-Morales et al. (2020) <sup>16</sup>	coarse poplar wood (PW)	0	0	146.3	7.0	10.0	8.6	- Fixed bed torrefaction at bench-scale (450g).	
		240	1	148.2	9.3	7.3	5.4	- Other tests performed like the previous work <sup>88</sup>	
		280	1	153.3	10.4	6.0	4.9		
	fine poplar wood (PW)	0	0	149.1	5.6	5.8	6.0		
		240	1	146.2	4.6	4.3	4.1		
		280	1	151.1	5.8	4.6	4.1		
Thevenon et al. (2021) <sup>89</sup>	mix of spruce/pine wood (SPW)	0	0	60	5	14.7	5.9	10	- Six-hearth furnace for torrefaction at pilot-scale (420kg).
		250	0.92	61	9.6	16.4	18.7	17.3	- Other tests performed: elemental analysis, calorific value, PSD, and shape factor.
Rajaonarivony et al. (2023) <sup>90</sup>	wheat straw (WS)	0	0	36.10					- Continuous screw reactor for torrefaction at pilot scale (2.92 kg/h). - Other tests performed: biochemical composition, AFM, TGA, NMR, calorific value, PSD, and shape factor.
		280	0.17	32.10					

522 Note: FFC<sup>a</sup> was measured at pre-consolidation loads of 2-2.7 kPa, FFC<sup>b</sup> at 5-5.3 kPa, FFC<sup>c</sup> at 7.9 kPa, and FFC<sup>d</sup> at 10-10.5 kPa.

523 Conag *et al.*<sup>82</sup> investigated the torrefaction of sugar cane bagasse of 0.56 mm using about  
524 9 g of material in a fixed bed reactor at different temperatures (*i.e.*, 250°C, 300°C, and 350°C)  
525 and residence times (*i.e.*, 15, 30, 45, 60, and 75 min) under minimized oxidative atmosphere.  
526 The authors evaluated the flowability via HR and CCI, indicating that the torrefied biomass had  
527 relatively lower indices than the raw biomass, meaning better flowability but also a worse  
528 compaction for these samples. The qualitative indication for the raw sample was “very poor”  
529 and “passable” for the torrefied powder. Another finding was that a torrefaction time beyond  
530 60 min did not further improve the fuel characteristics of torrefied biomass, in terms of energy  
531 density and high heating value.

532 Rago *et al.*<sup>83</sup> performed the torrefaction of waste textile fibers of about 9 mm mesh size  
533 at different temperatures (*i.e.*, 225°C, 250°C, 275°C, and 300°C) for 1 and 3 h in a batch reactor  
534 with about 65 g of material. The flowability was assessed via the Hausner ratio and  
535 compressibility index, generally showing a better flowability as the torrefaction severity (*i.e.*,  
536 higher temperature and/or higher treatment time) was increased. The initial fibers were highly  
537 fibrous and fluffy, corresponding to the highest bulk density compared to the torrefied material,  
538 to which the characteristics of a more powdery, less fluffy, and rather porous material were  
539 attributed.

540 Xu *et al.*<sup>84</sup> evaluated the flowability of four milled biomass powders (soybean straw, corn  
541 straw, rice straw, rice husk) before and after torrefaction in a lab-scale fixed bed reactor  
542 operated at 300°C for 45 min (amount of produced powder not indicated in the paper). The  
543 flowability was assessed via static angle of repose (fixed bed method) for two sieve size ranges  
544 of 250-380 µm and 380-550 µm. Overall, better flowability was observed for some torrefied  
545 samples (soybean and rice straws), particularly for the finer particles, while for others (rice  
546 husk, corn straw) the angle of repose remained almost identical even with the torrefaction  
547 pretreatment. Moreover, although torrefaction can help in improving the flowability, it cannot

548 change the relative relationship between the flowability of different biomass powders. Finally,  
549 through compressibility tests on raw milled biomass powders, the authors stated that the  
550 Hausner ratio/compressibility index cannot always characterize their flowability compared to  
551 the angle of repose, as a consequence of biomass nature (*i.e.*, specific internal structures,  
552 flexibility, and shape). Note that the compressibility tests were not evaluated for the torrefied  
553 sample.

554 Singh *et al.*<sup>85</sup> investigated the dry torrefaction of pigeon pea stalks of sieve sizes between  
555 0.7 and 1.2 mm under different temperatures (*i.e.*, 225°C, 250°C, and 275°C) and residence  
556 times (*i.e.*, 15, 30, and 45 min) using samples of about 6 g in a batch furnace. The authors  
557 verified a decrease in the solid yield with increasing torrefaction severity, which was defined  
558 by higher residence times and temperatures in the reactor. The flowability assessed via HR and  
559 CCI was improved by increasing the torrefaction severity, being classified as “very poor” for  
560 the raw sample and “poor” for the condition of 275°C and 45 min. Nevertheless, the authors  
561 state that a low temperature of 225°C at any of tested residence times had no effects for  
562 flowability improvement. Finally, the torrefied biomass showed a higher internal porosity and  
563 lower bulk density (about 18% lower than raw biomass), tended to absorb less water, and had  
564 reduced structural strength with more surface cracks that in turn helps in reducing crushing and  
565 compaction power consumption.

566 In a following paper, Singh *et al.*<sup>86</sup> evaluated the torrefaction of 8g eucalyptus wood of  
567 sieve sizes between 0.7 and 1.2 mm using the same reactor setup at similar temperatures (*i.e.*,  
568 220°C, 250°C, and 280°C) and residence times (*i.e.*, 20, 40, and 60 min). Similar conclusions  
569 were drawn from the experiments, consisting of an improvement of the flowability via HR and  
570 CCI as the torrefaction severity was increased. At the limits, the raw sample presented a “poor”  
571 flowability that was transformed into “fair” for the sample treated at 280°C and 60 min.

572 Singh *et al.*<sup>33</sup> experimentally evaluated the dry torrefaction in a fixed bed reactor, by using  
573 about 10g of gum Arabic wood powders in each test, with a sieve size from 0.7 to 1.2 mm. They  
574 evaluated various conditions for torrefaction temperature (from 220°C to 280°C), residence  
575 time (from 20 to 60 min) and heating rate (from 5°C to 15°C/min), aiming at obtaining optimum  
576 conditions for maximization of torrefied biomass high heat value and energy yield through  
577 statistical Response Surface Methodology (RSM). The flowability was evaluated via HR and  
578 CCI for the raw and torrefied sample of highest yield (*i.e.*, 252°C, 60 min, and 5°C/min),  
579 showing an improvement of flowability from “fair” to “good”, respectively.

580 In a following paper, Singh *et al.*<sup>87</sup> evaluated the torrefaction of gum Arabic wood at  
581 similar residence times (*i.e.*, 20, 40, and 60 min) and temperatures (*i.e.*, 220°C, 250°C, and  
582 280°C), obtaining the optimum tradeoff between higher heating value and energy yield for  
583 250°C and 40 min. The flowability was evaluated via static angle of repose, HR and CCI for  
584 the raw sample and three other torrefied ones at 40 min and all temperatures. There was a  
585 general improvement in the flowability indices, from the raw sample being classified as  
586 “passable” and the torrefied at the highest temperature as “good”. It is worth mentioning that  
587 the qualitative classification among different indices did not match perfectly, which is a  
588 common finding in the literature and agrees with the rationale from Section 3.2.1, illustrating  
589 that different flowability indices describe the flow behavior at different bulk flow conditions.  
590 Also noteworthy is the use of advanced physicochemical characterization techniques in these  
591 last two studies, such as FTIR, XRD, BET, and ICP-MS.

592 Pachón-Morales *et al.*<sup>88</sup> investigated the torrefaction of poplar and spruce wood chips of  
593 1.5 mm of particle diameter at three temperatures (*i.e.*, 220°C, 250°C, and 280°C) and two  
594 residence times (*i.e.*, 1 and 5 h) in a batch furnace containing about 15 g of material. Under  
595 similar torrefaction intensities, Spruce presented higher formation of fines (particles below 100  
596 µm) compared to poplar, which was attributed to distinct anatomical structures for the two



597 biomasses that lead to different fracture pathways. Besides, a gradual shifting of size and shape  
598 distributions towards finer and rounder particles was verified when increasing the torrefaction  
599 intensity. The flowability for all powders was evaluated with the Schulze ring shear tester at  
600 consolidations of 2, 5, and 10 kPa. A significant improvement of the flowability was verified  
601 as the torrefaction intensity was increased, from “cohesive” for the raw samples to nearly “free  
602 flowing” for the condition of 280°C during 5 h. Moreover, the authors provided a  
603 comprehensive discussion to state that the improvement of flowability was mostly related to  
604 the rounder shape and sharper surface of torrefied biomass, resulting from a loss of resilience  
605 of the fibrous structure of raw wood due to torrefaction, and not by the changes in the particle  
606 size or size span.

607 In a following study, Pachón-Morales *et al.*<sup>16</sup> focused on the torrefaction of poplar wood  
608 chips at 240°C and 280°C for 1 h in a batch furnace using samples of about 450 g. After grinding  
609 and sieving, the authors evaluated the flowability of raw and torrefied powders of two different  
610 particle-size distributions through techniques of shear cell and rotating drum, representatives of  
611 compacted and free-surface stress states, respectively. Overall, intense torrefaction improved  
612 the flow behavior of powders, both under free-surface and consolidated conditions. Fine, mildly  
613 torrefied powders showed the worst flow behavior, whereas the intensively torrefied coarse  
614 sample has the best flowability. Note that for some conditions the flowability of raw biomasses  
615 was better than the corresponding torrefied sample, indicating potentially that this pretreatment  
616 does not always improve the flow behavior. This highlights the need for systematically  
617 measuring the flow properties to draw best conclusions for ease handling and storage  
618 operations.

619 Thevenon *et al.*<sup>89</sup> investigated the effect of torrefaction at pilot scale for wood chips,  
620 consisting of a mix of 70% spruce and 30% pine, at the temperature of 250°C and residence  
621 time of 55 min. The raw sample contained a moisture content of 52% wet basis, which was

622 initially dried in a pilot scale belt dryer to about 18% wet basis, and subsequently torrefied in a  
623 continuous vertical six-hearth furnace (10 m in height and 1.82 m in diameter) to produce 420  
624 kg of torrefied wood. Both raw and torrefied wood chips were ground with a knife mill  
625 generating powders samples with mean sieve size of 662 and 264  $\mu\text{m}$ , respectively. Torrefaction  
626 not only reduced the grinding energy costs but also decreased by half the particle diameter,  
627 which is related with the weakening of the particle structure due to torrefaction. The flowability  
628 was evaluated with a rotating drum and Schulze shear cell representing bulk conditions of loose-  
629 dynamic and consolidated-static states, respectively. With respect to the dynamic avalanche  
630 angle, the raw and torrefied samples presented similar mean values of  $60^\circ$  and  $61^\circ$ , respectively,  
631 however, the first had a pronounced erratic flow behavior evidenced by standard deviation of  
632  $38^\circ$ , while the flow for the torrefied sample was more homogeneous with a deviation of  $8^\circ$ .  
633 Concerning the shear tests, the raw sample was classified as a “easy-flowing” material, while  
634 the torrefied was “free-flowing”, with the flowability change majorly attributed to the  
635 smoothing of particle surfaces as the aspect ratio of the particles remained almost stable in their  
636 work. Overall, the authors concluded that there was an improvement in the flowability, under  
637 both loose and consolidated states, due to the torrefaction treatment.

638         Rajaonarivony *et al.*<sup>90</sup> performed the torrefaction of wheat straw in a pilot scale screw  
639 reactor at  $220^\circ\text{C}$  and  $280^\circ\text{C}$ , feed rate of 2.92 kg/h, and residence time of 10 min. Torrefied and  
640 non-torrefied biomass were ground in a stirred ball mill to obtain fine powders in the range of  
641 3 to 120  $\mu\text{m}$ . Torrefaction at the low temperature of  $220^\circ\text{C}$  did not result in significant changes  
642 neither in biomass physicochemical and structural properties nor in their grindability.  
643 Nevertheless, at  $280^\circ\text{C}$  there was 50% better grindability compared with the raw sample, but  
644 also with higher tendency for agglomeration of fines, forming hard agglomerates in the range  
645 of 200-600  $\mu\text{m}$ . The agglomeration phenomena originated by torrefaction was attributed to  
646 modifications of particle surface properties that make certain chemical groups more accessible

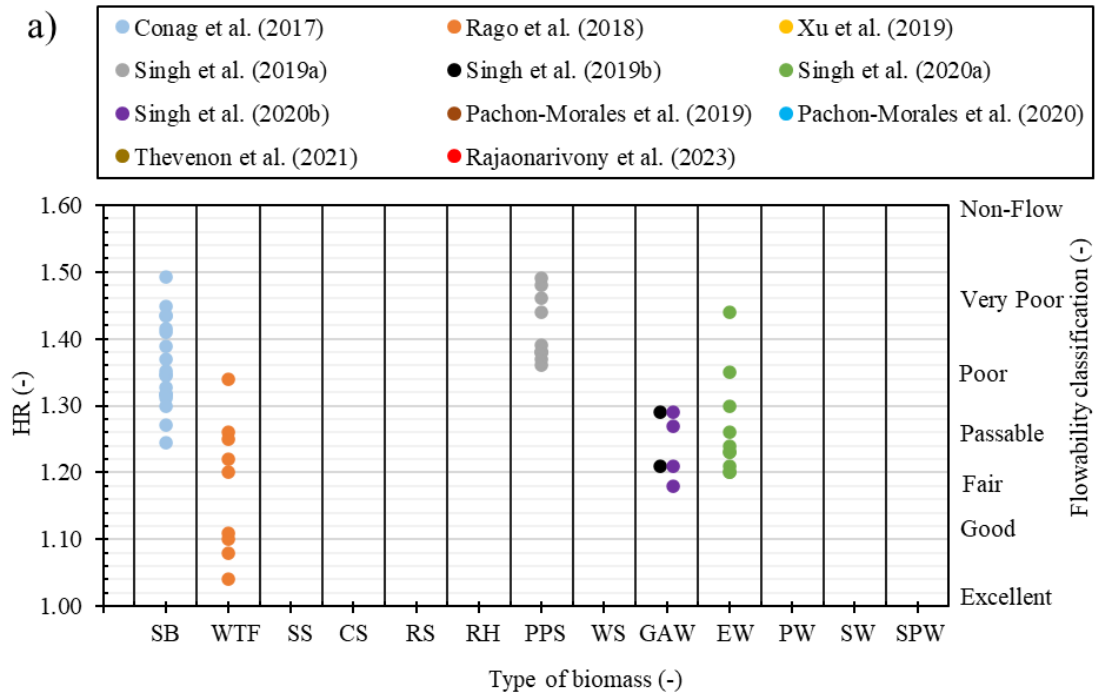
647 to form stronger interactions with others, but it requires further investigation. The flowability  
648 was evaluated through compressibility index with applied pressure of 15 kPa and with FT4  
649 rheometer for cohesion measurement. Overall, the flowability was improved from “very poor”  
650 for the raw sample to “poor” for the torrefied powder at 280°C, while the cohesion was also  
651 reduced by half in the latter. Torrefaction process makes straw particles more brittle, which  
652 decreases their deformability and compressibility of the powder. Moreover, the authors state  
653 that even a small improvement of the compressibility index (measured at quasi-static  
654 conditions) would represent huge gains on powder handling as they can be associated with  
655 significantly less mechanical forces to initiate the movement of particles. Besides, the cohesion  
656 reduction in the torrefied powder was interestingly associated with the degradation of  
657 arabinoxylans, a non-cellulosic component responsible for interparticle interactions of cohesive  
658 nature.

659

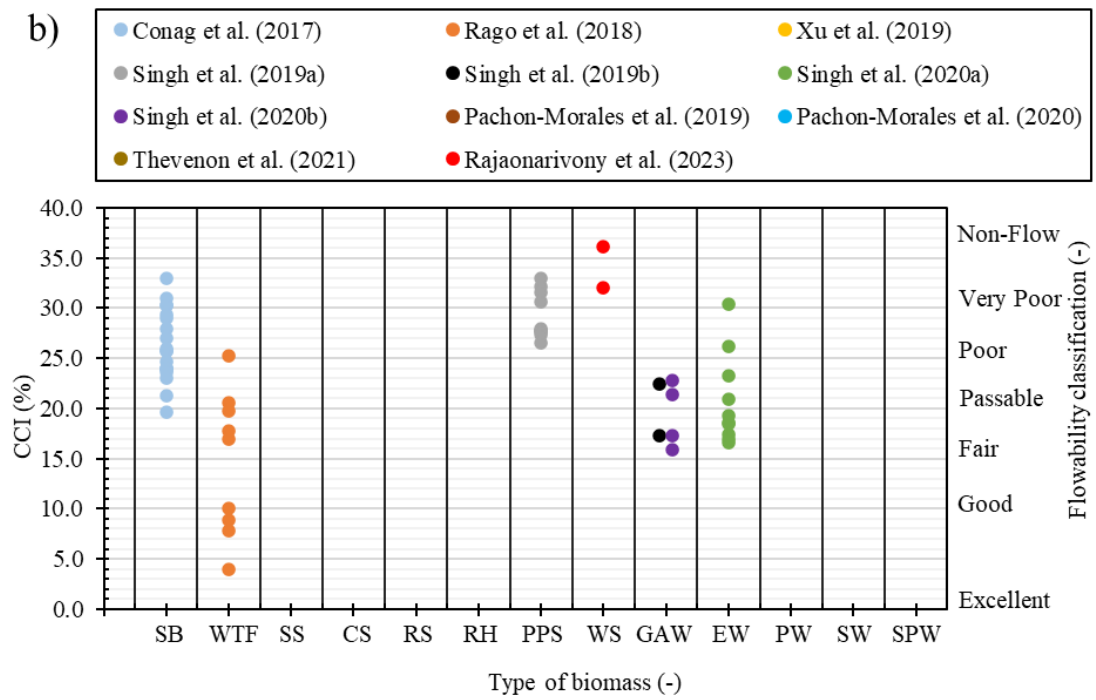
### 660 *3.3.3 Grouped flowability data and results discussion*

661 In this section, the flowability data gathered in Table 1 are discussed in more detail to  
662 reveal the current status of the literature and visualize the gaps that need future studies. The  
663 flowability indices considered were the HR, CCI,  $\alpha$ ,  $\beta$ , and FFC at different pre-consolidation  
664 loads, for which an overview was done in Section 3.3.1. Dispersion plots were produced for  
665 each flowability index as a function of the investigated biomasses, a total of 13 feedstocks. In  
666 Figure 5, the data points are colored based on the 11 literature references for flowability, while  
667 no occurrence of data points for a given biomass means that it has not been characterized with  
668 the corresponding flowability technique. At the right axis, the commonly used flowability  
669 classifications from the literature were applied for HR<sup>52</sup>, CCI<sup>53</sup>,  $\alpha$ <sup>76</sup>, and FFC<sup>67</sup>. Note that a  
670 classification for  $\beta$  is not yet available in the literature, as detailed in Section 3.3.1.

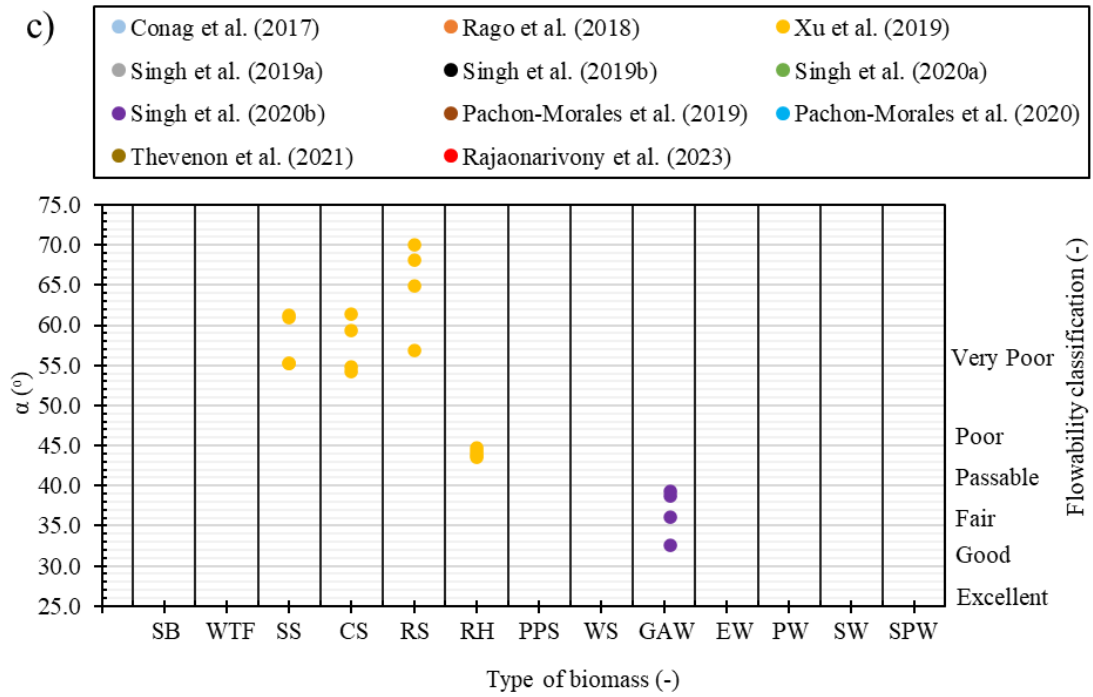
671



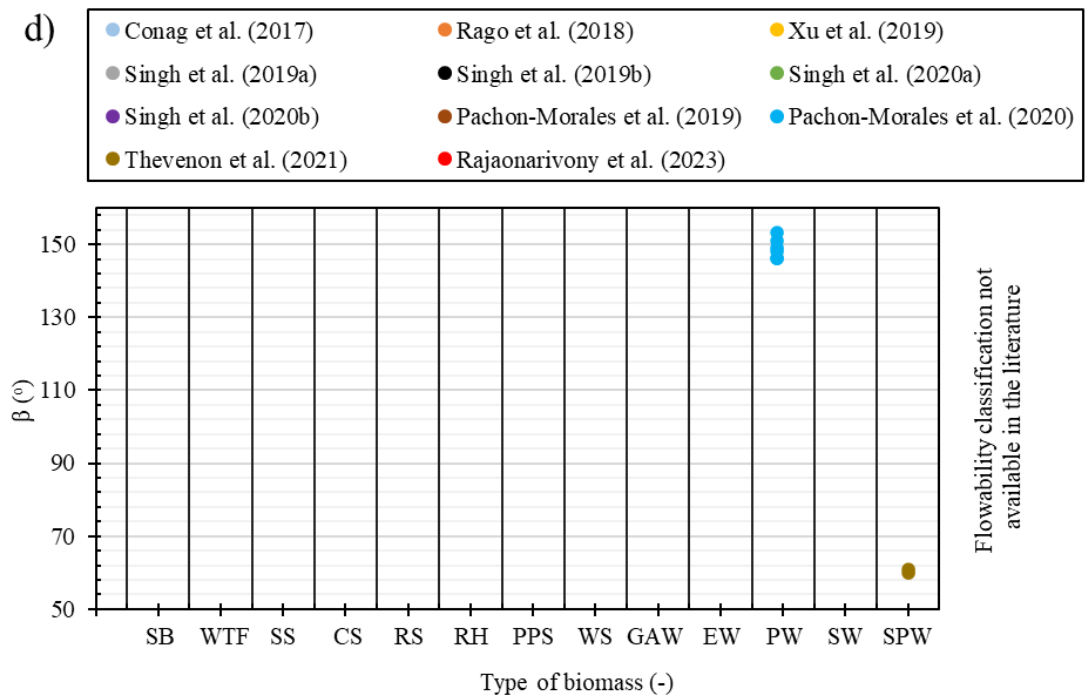
672



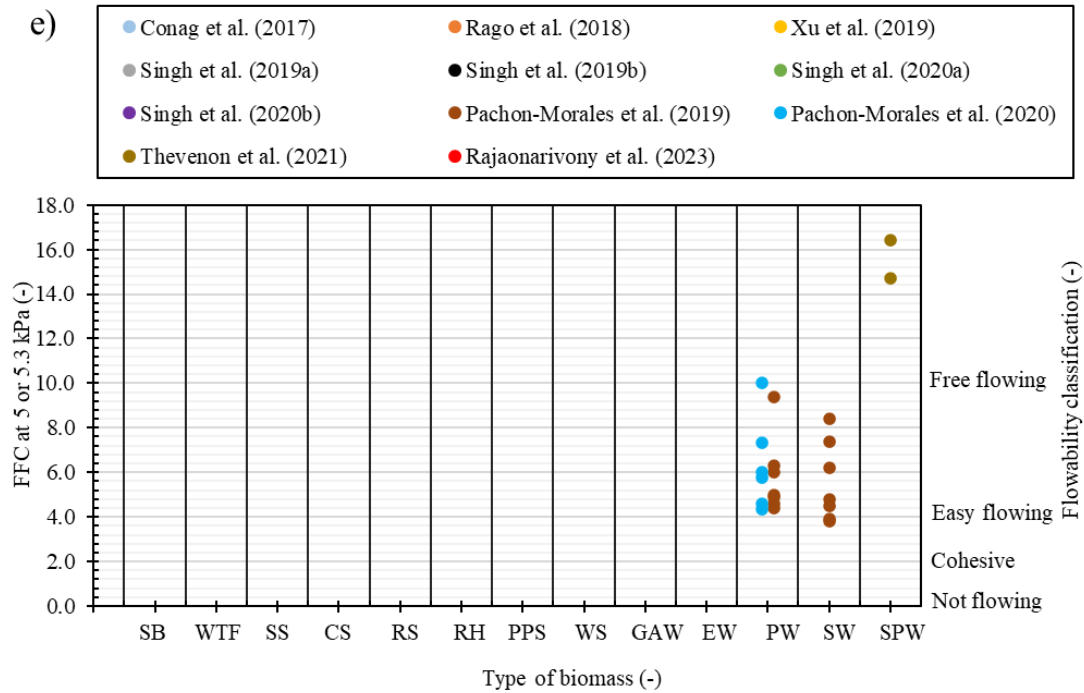
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674



675



676 Figure 5. Flowability indices as a function of different investigated biomass in the literature:  
 677 a) HR, b) CCI, c)  $\alpha$  d)  $\beta$ , and e) FFC. The points are colored based on the literature reference.  
 678  
 679 No occurrence of data points for a given biomass means that it has not been characterized  
 680 with the corresponding technique.  
 681

682 Overall, the classical indices derived from powder bed compaction, HR, and CCI, were  
 683 those mostly used to assess raw and torrefied biomass flowability. Data dispersion in Figures  
 684 5a and 5b revealed similar trends for both indices including similar flowability classification,  
 685 as expected, since both are based on the same experimental unit and raw data. In total, 5  
 686 feedstocks were characterized via HR with only one (*i.e.*, gum arabic wood) being evaluated  
 687 by two different studies. Similarly, for the CCI, 6 feedstocks were assessed with only gum  
 688 arabic wood being evaluated by two papers. Depending on the torrefaction treatment severity  
 689 and feedstock, HR ranged from 1.10 (excellent flowability) to 1.49 (very poor). Similarly, CCI  
 690 ranged from 3.9% (excellent) to 36.1% (very poor).

691 In Figure 5c, the static angle of repose was measured for 5 feedstocks in two literature  
 692 papers, highlighting a considerable variation of flowability classification, from 32.5° (good  
 693 flowability) to 70° (very poor), depending on the biomass sample and pre-treatment intensity.

694 It is worth noting that both papers used different techniques to measure the angle of repose,  
695 consisting of fixed diameter and fixed height, which makes a direct comparison difficult and  
696 establishes a warning for future studies. A similar comment can be attributed to Figure 5d, in  
697 which only two papers considered the rotating drum technique, and yet the criteria used for  
698 measuring the dynamic angle of repose was different among them, which explains the scattered  
699 data and prevents comparison of results.

700 In Figure 5e, shear cells were used to assess the flowability of raw and torrefied wood  
701 powders in three papers, for which the FFC (at a pre-consolidation load around 5kPa) ranged  
702 from 3.8 (easy-flowing) to 16.4 (free-flowing). The data trends for FFC under different  
703 consolidation loads can be observed in Table S3 (Supplementary Information). One of the  
704 advantages to use of shear cells for flowability classification is that the parameters for silo  
705 design, such as angles of internal friction and particle wall, can be made available from the  
706 same set of experiments. Coincidentally, all papers used the Schulze ring shear tester for the  
707 flowability data from Figure 5e; hence, it would be interesting in the future to compare these  
708 results with other commercial implementations of shear cells available in the literature. An  
709 illustration for this comparison was done previously for raw biomass.<sup>49</sup>

710 Based on the previous flowability dataset (Table 1 and Fig. 5) and the overview of current  
711 works associated with biomass torrefaction (Section 3.3.2), we found the following gaps and  
712 potential fields for future work:

- 713 - None of the studies on biomass torrefaction provided a complete picture of flowability  
714 using all available techniques, corresponding to different bulk flow conditions. In  
715 addition, coupling this with a full characterization of physicochemical, thermal, and  
716 bulk properties and techno-economic feasibility studies is pressing to improve the  
717 performance of thermochemical processes and the circular economy of biomass and

718 biomass residues. Noteworthy is the use of advanced characterization techniques that  
719 were employed in only 3 out of 11 works (see Table 1 for details).

720 - The comparison of results between the 11 studies was challenging as the flowability  
721 measurement techniques were not always the same, nor were the torrefaction  
722 conditions or the biomass type (which also implies quite different physicochemical  
723 and bulk properties: size, shape, surface roughness, bulk density, etc.). Consequently,  
724 current literature results are difficult to be applied elsewhere. Further works are  
725 needed, under similar flowability testing conditions and under a determined range of  
726 biomass properties, in order to obtain more sound and broad conclusions that allow  
727 for the establishment of standardized protocols for handling heterogeneous  
728 feedstocks, such as raw/torrefied biomass, at different process scales.

729 - None of the studies investigated the effect of electrostatic forces or evaluated in-depth  
730 interlocking effects or cohesion induced by moisture absorption, which prevents  
731 further discussions in this regard. The current focus of work was to report an  
732 improvement or not in the flowability, for very specific combinations of a given  
733 biomass and torrefaction condition. Further works centered on raw/torrefied biomass  
734 flowability assessment under higher intensity of electrostatic, cohesive & interlocking  
735 forces would be useful, not only for the bulk solids handling research field but also  
736 for more assertive applications with heterogeneous feedstocks.

737 - Most of the studies in the literature (9 out of 11) were based on small-bench  
738 torrefaction units, producing only a few grams of torrefied biomass. This is generally  
739 a limiting amount of material for complete assessment of flowability at different bulk  
740 conditions, as well as to evaluate other important handling aspects at industrial scale,  
741 such as bulk solid segregation, quality of fluidization, and so on. Thus, we suggest



742 this as a scope for future work aiming at improving the success of biomass torrefaction  
743 at different unit scales.

744

#### 745 3.3.4 Description of selected papers for fluidization

746 Rousset *et al.*<sup>91</sup> investigated the fluidization characteristics of wood chips of three size  
747 distributions, corresponding to raw and torrefied biomass at 200°C and 15 min. Overall, a  
748 higher pressure drop was observed for the raw biomass samples at the same gas velocity, which  
749 was attributed to the weakening of the torrefied sample. In a subsequent work, Rousset *et al.*<sup>92</sup>  
750 evaluated the grindability by fluidization of three wood chips samples with different size  
751 distributions, before and after torrefaction for 15 min at 210°C, 240°C, and 270°C. The  
752 fluidization tests were performed with 450 g of material and for 20 min at an increasing gas  
753 velocity up to 3 m/s. With respect to the raw samples, only big particles higher than 1.6 mm  
754 showed a reduction in size by fluidization. On the other hand, torrefaction had a significant  
755 impact on particle grindability by fluidization for all sizes from 0.28 to 1.6 mm. The size  
756 reduction was also correlated with the torrefaction severity, with higher treatment temperatures  
757 originating finer particles. Nevertheless, further studies are needed in this field to show the pros  
758 and cons of biomass torrefaction for fluidized bed technologies, particularly by studying more  
759 complicated biomass residues (*e.g.*, refuse-derived fuel or municipal waste) and by  
760 investigating other important phenomena in fluidization, such as gas-solid contacting, particle  
761 segregation, particle attrition, and particle entrainment.

762

#### 763 3.4 Current applications of torrefied biomass and future perspectives

764 Not all retrieved studies on the flowability and fluidization of torrefied biomass  
765 highlighted the potential applications of their findings. Most of these studies primarily focused

766 on enhancing the energy potential of biomass. Proper physical and bulk characterization is  
767 essential to improve the handling and stability of torrefied biomass, an important step to scale  
768 up the experiments to field applications. Additionally, there are environmental benefits of  
769 torrefied biomass, including soil amendments and remediation<sup>93</sup>, but the potential risks and  
770 limitations of both the torrefaction process and torrefied biomass itself should also be explored.

771 The substitution of coal and other fossil fuels with torrefied biomass presents a  
772 compelling alternative for mitigating the environmental impacts associated with energy  
773 production. Co-firing torrefied biomass alongside coal, for example, has been shown to  
774 significantly decrease greenhouse gas emissions from power plants.<sup>94</sup> Torrefied biomass  
775 typically exhibits higher heating values and calorific potentials than those of raw biomass,<sup>95</sup>  
776 making it a more efficient energy source. This enhancement in energy density not only  
777 improves combustion efficiency but also expands the range of applications for biomass-derived  
778 fuels in various industrial processes, which can contribute as a solution in transitioning towards  
779 cleaner energy systems. In addition, biomass torrefaction offers an alternative method for  
780 repurposing agricultural residues, such as the energy densification of sugarcane bagasse, as  
781 demonstrated in the study by Conag *et al.*<sup>82</sup>, and the improvements in biomass powder  
782 flowability for different biomasses (soybean straw, corn straw, rice straw, and rice husk), as  
783 explored by Xu *et al.*<sup>84</sup>.

784 Torrefied biomass offers several advantages over raw biomass, particularly in terms of  
785 transportation, storage, and densification.<sup>96</sup> Due to its reduced moisture content and improved  
786 bulk and flow properties resulting from the torrefaction process, such as increased energy  
787 density and enhanced grindability, torrefied biomass exhibits higher stability during long-  
788 distance transportation. This stability minimizes the risk of particle degradation and ensures  
789 consistent quality upon arrival at its destination, making it more economically viable and  
790 practical for large-scale energy production facilities. The densification of torrefied biomass into

791 pellets or briquettes becomes more efficient and cost-effective, further streamlining handling  
792 and storage processes. These characteristics improve logistical efficiency and facilitate the  
793 integration of biomass-based fuels into existing energy infrastructure, promoting sustainable  
794 energy solutions with reduced environmental impact. In addition, the evaluation of torrefaction  
795 pre-treatment on pellet durability with and without binders is also an important topic for  
796 efficient feeding of these materials to reactors that needs further investigation, particularly to  
797 feed more materials while avoiding issues of pellet breakage or segregation.<sup>97</sup>

798         The application of torrefied biomass can significantly enhance soil properties, offering  
799 benefits such as increased water-holding capacity and improved nutrient availability.<sup>93,98</sup> While  
800 torrefaction increases the hydrophobicity of biomass, water-holding capacity is enhanced due  
801 to the formation of a porous structure of micro- and mesopores (interporosity) and within the  
802 torrefied biomass particles (intraporosity)<sup>99,100</sup>. However, as explored by Edeh and Mašek<sup>101</sup>  
803 biochar hydrophobicity increases with decreasing particle size, leading to a reduction in water  
804 retention capacity; they recommend using hydrophilic biochars with high intraporosity to  
805 enhance water retention in coarse soils. In this sense, torrefied biomass can reduce hydraulic  
806 conductivity, decrease water losses due to evaporation, and make water more available for plant  
807 uptake<sup>102</sup>. This enhanced water retention capability not only reduces irrigation needs but also  
808 promotes better moisture management<sup>100</sup>, particularly in arid or drought-prone regions.

809         Torrefied biomass releases nutrients slowly over time as it decomposes, providing a  
810 steady supply of essential elements like nitrogen, phosphorus, and potassium to support plant  
811 growth.<sup>92</sup> These nutrients act as natural fertilizers, enhancing soil fertility and productivity  
812 without the immediate risk of leaching or runoff, thereby contributing to sustainable agricultural  
813 practices. In contaminated sites, torrefied biomass can be a potential cost-effective adsorbent  
814 for the retention and sorption of contaminants.<sup>103,104</sup> Its ability to effectively bind pollutants  
815 makes it promising for the remediation and reuse of organic wastes and residues. Despite these

816 advantages, the scalability of these applications requires thorough investigation, as well as an  
817 understanding of the long-term effects of torrefied biomass application in soils. Potential  
818 desorption of contaminants over time can hinder the environmental sustainability and  
819 effectiveness of remediation strategies. Further research is needed to explore these aspects,  
820 ensuring the proper application of torrefied biomass in environmental remediation.

821 The utilization of torrefied biomass, while offering several environmental benefits, also  
822 presents potential risks that require critical assessment. The selection of the feedstock for  
823 producing the torrefied biomass should consider location factors and logistics, considering  
824 biomass residues largely produced and available near the processing units. The energy input  
825 required for the torrefaction process should be considered for assessing the carbon savings  
826 achieved during combustion. Improper disposal or land application of ash could lead to soil  
827 contamination and nutrient imbalances, impacting local ecosystems and agricultural  
828 productivity. Furthermore, the long-term impacts of torrefied biomass application in soil  
829 remediation and agriculture still need further research, as the torrefied biomass itself can be  
830 toxic for several organisms.<sup>105</sup>

831

#### 832 **4 Final considerations**

833 From this systematic review on biomass torrefaction, it was observed that most papers  
834 focused on determining the chemical and thermal properties of raw and torrefied biomass (62%)  
835 to maximize its energy density and solids yield. In contrast, only some studies (8%) presented  
836 a complete characterization of physical and bulk properties, and less than 2% investigated  
837 flowability or fluidization characteristics for the raw and torrefied biomass, which is critical for  
838 the reliable operation of processes at the pilot to industrial scales. This highlights the need for  
839 future research on the flowability of raw and torrefied biomass, with the goal of obtaining more

840 sound and broad conclusions for the flow behavior of such heterogeneous materials,  
841 culminating in the development of standardized protocols to improve biomass handling and  
842 processing at an industrial scale. Some considerations for future studies are summarized below:

843 - No paper has performed a complete characterization of raw and torrefied biomass  
844 encompassing physicochemical, thermal, bulk, and flow properties, a surprising feature  
845 that should be covered in future studies.

846 - Most experimental works were based on the torrefaction of limited amounts of biomass,  
847 on the order of a few grams, which can be limiting for extrapolation or interpretation of  
848 flowability results to industrial scale process conditions, particularly because at this  
849 latter scale other bulk solids challenges that can prevent powder flow may arise, such  
850 as more pronounced powder segregation, effect of powder bed resistances, effect of  
851 pipeline defects and wear, etc. Therefore, future studies should assess bulk and flow  
852 properties from large-scale torrefaction units and compare them with small-scale ones.

853 - Concerning the flowability characterization, current works focused more on simple  
854 classical methods based on powder compressibility or static angle of repose, while very  
855 few employed shear cells or rotating drums. Moreover, none among the (few) studies  
856 on biomass torrefaction provided a full picture of flowability characterization using all  
857 the available techniques. Thus, future studies could cross-check different flowability  
858 techniques for a range of raw and torrefied biomass and verify the reproducibility of  
859 results by using different equipment brands of a similar measurement technique.

860 - Currently, the studies focused more on the characterization of wood and agro-industrial  
861 biomass wastes, however, in the future more heterogeneous and complex wastes, such  
862 as municipal wastes or refuse-derived fuels, should be investigated concerning  
863 chemical, physical, bulk, and flow properties, aiming at proposing successful solutions  
864 for thermochemical valorization of such materials.

865 **Disclosure statement**

866 The authors declare no competing financial interest.

867

868 **Acknowledgments**

869 A.P.O. has a postdoctoral grant from the São Paulo Research Foundation (FAPESP, Process  
870 2021/14789-1).

871

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883 **Chayene Gonçalves Anchieta** specializes in materials characterization for energy applications.  
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899

900 **Authors contributions**

901 **Lucas Massaro Sousa:** Conceptualization, Methodology, Formal analysis, Investigation,  
902 Writing – Original draft, Reviewing and Editing; Supervision; Project Administration; **Allan**  
903 **Pretti Ogura:** Methodology, Formal analysis, Investigation, Writing – Original draft,  
904 Reviewing and Editing; **Chayene Gonçalves Anchieta:** Methodology, Formal analysis,  
905 Investigation, Writing – Original draft, Reviewing and Editing; **Mathieu Morin:** Writing –  
906 Reviewing and Editing; **Nicholas Slongo Canabarro:** Conceptualization, Methodology,  
907 Formal analysis, Investigation, Writing – Original draft, Reviewing and Editing.

908

909 **Abbreviations**

910  $\alpha$ : static angle of repose;  $\beta$ : angle of repose; AFM: atomic force measurement; ASTM:  
911 American Society for Testing and Materials; AWL: Anhydrous Weight Loss; CCI: Carr's  
912 compressibility index; CHNS/O: carbon, hydrogen, nitrogen, sulfur, and oxygen ratio; CT:  
913 computed tomography; FFC: flow function coefficient; FTIR: Fourier Transform Infrared  
914 Spectroscopy analysis; NMR: nuclear magnetic resonance; H/C: hydrogen/carbon ratio; HR:  
915 Hausner ratio; ICP-MS: inductively coupled plasma-mass spectrometer O/C: oxygen/carbon  
916 ratio; PRISMA: Preferred Reporting Items for Systematic Reviews and Meta-Analyses; RSM:  
917 Response Surface Methodology; SAXS: X-ray-based synchrotron-based scattering; SEM:  
918 scanning electron microscopy; TGA: Thermogravimetric analysis; XPS: X-ray Photoelectron  
919 Spectroscopy; XRD: X-ray diffraction analysis; XRF: X-ray fluorescence.

920

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